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NUCLEAR ORIENTATION STUDIES OF RARE-EARTH METALS

by

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ABSTRACT

The angular distributions of gamma rays from ^{166m}Ho and ^{160}Tb aligned at low temperatures in, respectively, Ho metal and Tb metal have been measured. Large hyperfine splittings, expected for the rare earths, have been deduced from the temperature dependence of the gamma-ray anisotropies. Both samples show a macroscopic magnetic anisotropy which is not consistent with an interpretation in terms of a randomly oriented polycrystalline structure.

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The radioactive decay of oriented nuclei has become a successful and widely used technique for studies of both nuclear spectroscopy and hyperfine interactions. Such nuclear properties as spin assignments, magnetic dipole and electric quadrupole moments, and radiation multipolarities can be deduced from these experiments. Moreover, nuclei with known spectroscopic properties can be placed in different environments as probes of the interaction of nuclear moments with the electromagnetic fields of the host material. This technique has not been widely used for nuclear reaction studies. In part this stems from the necessity of preparing the desired nuclear species as a dilute impurity in a ferromagnetic host; in this case nuclear reactions with the host material would likely overwhelm any effects associated with the impurity. A further difficulty is the uncertainty in the actual degree of nuclear polarization resulting from the heating of the sample by the incident beam.

Rather than use a dilute impurity for such experiments, it would be preferable to use a material which is itself ferromagnetic and which presents interesting challenges for the study of both nuclear physics and hyperfine interactions. Such materials are the rare-earth metals, which are ferromagnetic at low temperatures and which can be used by the nuclear physicist to study a number of interesting nuclear properties and the systematic variation of such properties over the entire range of the rare earths, whose nuclear structure is characterized by large permanent quadrupole deformations.

As a preliminary step in evaluating the feasibility of rare earth polarized targets, we have investigated the angular distributions of gamma-rays emitted following the radioactive decays of rare-earth nuclei

produced in situ by neutron activation of bulk polycrystalline rare-earth metal samples. We have chosen for these studies the metals Ho and Tb. The expected hyperfine magnetic fields in these materials are sufficiently large (740 tesla for Ho, 314 tesla for Tb)¹ that the nuclear polarization is effectively saturated at the low temperatures easily attainable in the laboratory. The expected magnetic hyperfine splittings are 130 mK for ^{166m}Ho and 65 mK for ¹⁶⁰Tb.

Radioactive samples of ^{166m}Ho and ¹⁶⁰Tb were produced by neutron activation of polycrystalline samples of Ho and Tb metals. The Ho was in the form of a flat plate of thickness 1 mm and area 1 cm²; the Tb sample was a hemispherical button of radius about 3mm. In separate experiments the samples were soldered to the cold finger of a ³He-⁴He dilution refrigerator along with a ⁵⁴Mn in Fe thermometer. A split-pair superconducting coil was capable of supplying an external magnetic field of 4.5 tesla at the location of the sample. Gamma-rays were observed at 0° and 90° relative to the field direction using a pair of Ge(Li) detectors. Thermometry at the low temperatures (< 0.05 K) was done with the ⁵⁴Mn; at the higher temperatures calibrated resistance thermometers were used. The angular distributions, normalized by the high-temperature (4 K) counting rates, were analyzed with the usual expression

$$W(\theta) = 1 + \alpha_2 P_2(\cos \theta) + \alpha_4 P_4(\cos \theta) \quad (1)$$

where

$$\alpha_k = Q_k B_k U_k A_k \quad (2)$$

Since the goal of the present report is the interpretation of the alignment parameters B_k , we have chosen γ -rays from the respective decays whose nuclear properties are sufficiently well known that A_k and U_k can be calculated. For this purpose the 712-keV γ -ray emitted by ^{166m}Ho and the 299-keV γ -ray from ^{160}Tb were chosen. Both are essentially pure E1 transitions which follow allowed $\Delta J = 1$ beta decays. Complete angular distributions for both decay schemes, including deduced nuclear spectroscopic information (spins and multipole mixing ratios), will be presented in forthcoming reports.

With a magnetic field of 2.0 tesla applied in the plane of the sample, the ^{166m}Ho 712-keV anisotropy was well saturated at the ultimate temperature of 20 mK, but the saturation anisotropies, $W(0^0) - 1 = 0.350 \pm 0.005$ and $W(90^0) = -0.173 \pm 0.002$, are considerably smaller than the expected maximum values for a single crystal sample, 0.494 and -0.247, respectively.² This reduction has been interpreted by Marshak et al.³ as originating in part from incomplete saturation of the bulk holmium magnetization and in part from non-alignment of the local moment with the applied field in the polycrystalline sample. In the hexagonal crystalline structure of holmium, the local moment is canted out of the basal plane; the angle between the local moment and the basal plane varies with the angle between the c-axis and the direction of the applied field. The calculation of the reduction in anisotropy then requires a geometrical averaging over the array of microcrystals, which in turn requires some assumptions regarding the nature and orientation of the individual crystals. Following the approach of Marshak et al.³, we assume our sample to be composed of many randomly oriented, non-interacting single crystals whose dimensions are small with respect to the dimensions of the sample. Carrying

out the geometrical averaging over the sample, we find that the anisotropy is expected to be reduced to about 56% of its theoretical maximum; however, the observed anisotropies are reduced only to 70-75% of maximum. (Increasing the applied field to 4.0 tesla increased the anisotropy by about 5%.) A more stringent test of this model is to measure the anisotropy when the external field is applied perpendicular to the plane of the sample; in the case of small random microcrystals, we would expect essentially the same anisotropies independent of the direction of the field. However, with 4.0 Tesla applied field (which corresponds roughly to 2.0 Tesla applied field for the in-plane magnetization, owing to the large demagnetizing field in this poor geometry) the saturation anisotropies were $W(0^\circ) - 1 = 0.070 \pm 0.010$ and $W(90^\circ) - 1 = -0.045 \pm 0.004$. The immediate conclusion is that the random microcrystal model fails most likely because the individual crystals are not small with respect to the sample dimensions and are most probably aligned with the plane of the sample rather than randomly distributed. However, arbitrary distributions in the orientation and size of the individual crystals leave too many free parameters to make a meaningful fit to the data, and further information on the properties of the sample is required.

The Tb anisotropy shows similar properties. With the magnetic field parallel to the base of the hemisphere, the 299-keV anisotropy is fairly well saturated at 20 mK, with $W(0^\circ) - 1 = -0.269 \pm 0.003$ and $W(90^\circ) - 1 = 0.131 \pm 0.001$. (These are measured at 2.0 tesla applied field; at 4.0 tesla they are about 2% larger.) The reduction in anisotropy, as compared with the maximum value at saturation, is more serious than was the case with Ho; the measured values are 55% of the expected maximum. However, in this case when the sample was turned so that the applied field was perpendicular to the base

of the hemisphere, the observed anisotropy was less than 0.01. Although the source of this effect is not certain, a possible explanation is that the sample consists primarily of one large crystal or perhaps a few smaller (but still macroscopic) crystals, aligned so that the symmetry axis of the hemisphere is a hard direction for magnetization.

Even though the interpretation of the measured gamma-ray anisotropies is not yet clear, the present work has shown that large degrees of nuclear alignment can be obtained for bulk polycrystalline rare earth metals, and thus the use of such as targets for nuclear reaction experiments is possible. The degree of alignment obtained for Ho is less than that obtained for a Ho single crystal,² but exceeds the alignment obtained when Ho is prepared as a dilute impurity in gold⁴ or in a single crystal of neodymium ethyl sulfate.⁵ The Tb alignment exceeds that previously obtained for Tb impurities in Au⁶ and Gd.⁷

As a practical application a pure material such as Ho or Tb could be neutron activated and used as an aligned target for a nuclear reaction experiment. The angular distribution of the decay gamma rays emitted from the produced radioactivity could then serve as a reliable, convenient and continuous monitor of the degree of nuclear alignment of the target.

Since the gamma-ray angular distributions only serve to measure the even-order alignment parameters B_2 and B_4 , more information is needed for reaction experiments in which knowledge of the nuclear polarization (B_1 and B_3) is required. It is at this point that an understanding of the magnetic structure of the sample becomes necessary. If we had a model that could be

used reliably to calculate the dependence of B_2 and B_4 on temperature and on the magnitude and orientation of the applied field, then we would expect that B_1 and B_3 could be calculated with similar reliability. A direct measurement of B_1 , such as from the circular polarization of the emitted gamma-rays, would be a useful independent test of such a model.

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